

How speed of sound measurements could bring constraints on the composition of Titan’s seas

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ABSTRACT

The hydrocarbons seas of Titan, discovered by Cassini/Huygens mission are among the most mysterious and interesting features of this moon. In the future, a possible dedicated planetary probe, will certainly measure the speed of sound in this cryogenic liquid, as it was planned in the case of a Huygens landing into a sea. Previous theoretical studies of such acoustic measurements were based on rather simple models, leading in some cases to unphysical situations. Employed in a vast body of chemical engineering works, the state of the art PC-SAFT model has been recently introduced in studies aimed at Titan. Here, I revisit the issue of the speed of sound in Titan’s liquids, in the light of this theory. I describe, in detail, the derivation of the speed of sound from the chosen equation of state and the potential limitations of the approach. To make estimations of the composition of a ternary liquid mixture $\text{N}_2:\text{CH}_4:\text{C}_2\text{H}_6$ from speed of sound measurements an original inversion algorithm is proposed. It is shown that 50 measures between 90 K and 100 K are enough to ensure an accuracy of the derived compositions better than 10%. The influence of the possible presence of propane is also investigated.

Key words: instrumentation: miscellaneous – methods: numerical – techniques: miscellaneous – planets and satellites: individual: Titan.

1 INTRODUCTION

Among a multitude of fascinating features, Titan, the main satellite of Saturn, is – with the Earth – the only body of the solar system bearing stable liquid phases at its surface. These hydrocarbon seas and lakes remain largely mysterious: they appear amazingly flat (Zebker et al. 2014), with unexplained reflectivity events (Hofgartner et al. 2014), while their precise chemical composition is not well known. Only the presence of ethane has been detected (Brown et al. 2008) and estimations performed with numerical models (Tan et al. 2013; Glein & Shock 2013; Cordier et al. 2009, 2013) somewhat disagree. Given their important role as reservoir in the hydrocarbons cycle, and because of their high exobiological potential (McKay & Smith 2005; Schulze-Makuch & Grinspoon 2005; Lunine 2010), these lakes/seas would be very interesting targets for an *in situ* exploration. Already mission concepts are studied: for instance the project TiME (*Titan Mare Explorer*, see Stofan et al.

2011) proposes a sea surface exploration with a boat while Lorenz et al. (2015) suggest a submarine. Both proposals incorporate an instrument that exploits the properties of sound propagation in liquids.

The idea of instruments based on acoustic measurements has been investigated and implemented. of Mars. Originally aimed for thunder detection, sound sensors have been deployed at the surface of Venus (Ksanfomality et al. 1986a,b). Finally, Lorenz (1999) has discussed what could be learned about outer planet atmospheres using acoustic properties. In the context of Titan, the *Huygens* probe was equipped with the instrument called *Acoustic Properties Investigation* (API), belonging to the *Surface Science Package*, which consisted of two units: API-V (Velocity of sound) and API-S (Sounding) (Svedhem et al. 2004). Even if this instrument had also capabilities for performing analysis in the atmosphere, it was mainly aimed at liquid phase investigations. Since *Huygens* landed on a dry region, no measurements were done in Titan’s surface liquid. However acoustic data collected during the descent allowed Hagermann et al. (2007) to derive constraints on

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the atmosphere composition.

Hagermann et al. (2005) did a conceptual work of how the measurements, acquired by *Huygens'* instruments in a cryogenic liquid, could give information regarding its composition. Among other physical quantities, they considered the speed of sound. Facing the lack of published speed of sound measurements, particularly for the hydrocarbons mixtures, they used an equation of fit. They validated the latter by comparison with the results given by the equation of state (hereafter EoS) published by Peng & Robinson (1976). They also employed data coming from the National Institute of Standards and Technology (NIST14). However, they did not give the full derivation of the computed speed of sound. In the context of depths sounding, Arvelo & Lorenz (2013) made computations based on the speed of sound in cryogenic liquids. Their work also involves the NIST14 database. Unfortunately, the obtained velocities exhibit discontinuities which are certainly not physical (see figure 3 of Arvelo & Lorenz 2013). In addition, these authors assumed that their adopted linear fit can be extrapolated to temperatures higher than 92.5 K. For all these reasons, I decided to explore the question of speed of sound in cryogenic liquids and to revisit its sensitivity to chemical composition. Successfully used in countless works of chemical engineering, and introduced in Titan research field by Tan et al. (2013), I have chosen to use the up-to-date, Helmholtz energy based theory, PC-SAFT¹. On average, PC-SAFT is more accurate than all other EoS, and particularly Peng-Robinson cubic EoS (see for instance Diamantonis et al. 2013; Annesini et al. 2014). Focusing on the speed of sound dependency with chemical composition, I describe a concept of a very simple instrument, similar to the API-V, but including an active temperature control of the probed liquid. In this paper, I emphasize the required accuracy and on the needed number of velocity of sound measurements; this, in a way as quantitative as possible. In addition, the exact derivation of the speed of sound from PC-SAFT quantities is presented in detail. The system is not thought to compete with an instrument as accurate as a mass spectrometer, but it would be very useful in the case of a failure of such an high precision sensors. In Sect. 2 I discuss the principle of the speed of sound measurements. Sect. 3 is devoted to the thermodynamical computations and model description. In Sect. 4, the inversion algorithm is described and the sensibility of results to experimental conditions is discussed. Final remarks and conclusion will be made in Sect. 5.

2 THE PRINCIPLE OF THE MEASUREMENTS

If the bulk composition of Titan's lakes contains only three compounds, characterizing the composition can be achieved if we know the mole fraction of nitrogen x_{N_2} and the ratio $r_{46} = x_{CH_4}/x_{C_2H_6}$, where x_{CH_4} and $x_{C_2H_6}$ are respectively the mole fractions of methane and ethane. As shown, in principle, by Hagermann et al. (2005), an appropriate set

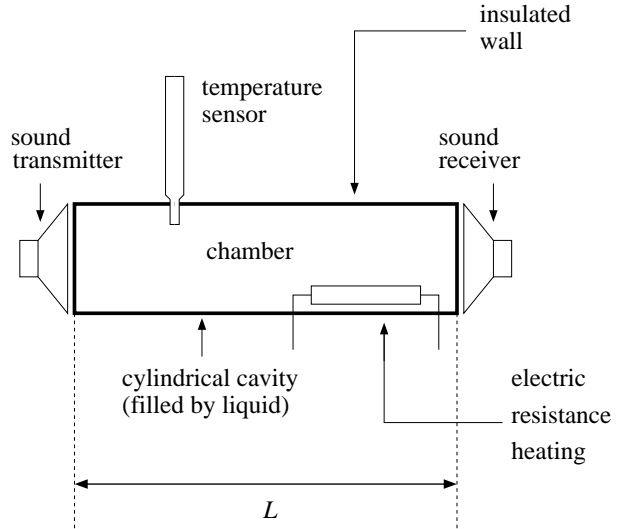


Figure 1. Sketch of the device that could be used to measure the speed of sound in the liquid of a Titan's lake. The temperature control of the liquid is ensured by means of temperature sensors and an electric heating system.

of physical property measurements can be used as an indicator of the chemical composition of the considered liquids. Following this idea, the measurements of two independent values of the speed of sound u in a liquid could be used – at least formally – to determine the values of x_{N_2} and r_{46} . These measurements could be performed for two different temperatures, and more extended set of measurements will bring stronger constraints on the composition. If u_1 and u_2 are respectively the determined sound velocity at temperature T_1 and T_2 , one has to solve the set of equations (1).

$$\begin{aligned} u(x_{N_2}, x_{CH_4}, x_{C_2H_6}, T_1) &= u_1 \\ u(x_{N_2}, x_{CH_4}, x_{C_2H_6}, T_2) &= u_2 \end{aligned} \quad (1)$$

This assumes that the liquid mixture does not evaporate at a temperature in the range $[T_1, T_2]$. Moreover the liquid has to be isolated from any vapor phase such as the local atmosphere. With more than two measurements, systems similar to (1), each corresponding to a couple $(T_i, T_j)_{i \neq j}$, can be resolved. The composition is then obtained by averaging the inferred mole fractions. In the frame of this approach, a law $u(x_i, T)$ has to be determined during the preparation of the space mission, by theoretical works and/or laboratory calibrations. One might conceive of a run of measurements in a passive mode, *i.e.* taking advantage of the natural variations of the surface temperature of the sea. Unfortunately, the expected amplitude of climatic temperature variations is of the order of a few kelvins by Titan's year (29.5 Earth years); these circumstances make these variations unusable for our purpose. Thus, an active system of temperature control is needed. A device inspired by the Kundt's tube (Kundt 1866) could be employed. Although it would be equipped by valves, such a system has the great advantage of a small number of mobile mechanical pieces. The Kundt's device is sketched in Fig. 1, enclosing the tube (that could also be called "chamber of measurement") an insulating wall prevents too high a heat flux towards ambient liquid, and stabilizes the temperature when the acoustic experiments are

¹ Perturbed-Chain Statistical Associating Fluid Theory

conducted. The probed liquid is heated by an electrical resistor whereas a device and/or a dedicated procedure ensure the homogeneity of the temperature in the chamber. The measurement can be done by determining the time taken by an ultrasonic pulse to travel the length of the tube between the emitter and the receiver. This is the operating principle achieved by API-V *Huygens* or “SOSO” (*TiME*). I emphasize that the tube could also be used in a way similar to what Kundt has done. Indeed, the tube is a cylindrical resonant cavity (of length L) The natural frequencies f_n of such a cavity obey the simple law given by Eq. (2)

$$f_n = n \frac{u}{2L} \quad (2)$$

(see Feynman et al. 1963). For a fixed cavity length L , the determination of only two consecutive resonant frequencies gives the speed of sound: $u = 2L(f_{n+1} - f_n)$. The measurement of a travel time is then replaced by a detection of resonance frequencies. The larger the sample of acquired data is, the more precise the value of the speed u is. An electronic unit dedicated to frequency generation and a signal receiver processing are required to complete the system. In addition, the liquid heating could be easily done with the help of an electrical resistor, while temperature measurements would be monitored by means of thermocouples. The global performances (*e.g.* number of measurements per unit of time) determine the actual accuracy achieved for the law $u(T)$.

In the next section, we discuss the precision needed for speed of sound determinations (or equivalently for frequencies) to get results useful for liquid composition. Concerning the range of temperature that would be used, I propose the interval from 90 K to 100 K. Indeed, the ground temperature of Titan's polar regions has been estimated to be ~ 90 K based on near-surface brightness temperature measurements (Jennings et al. 2009). In addition, methane has its boiling point at 111.2 K. It is surely technically easier to heat the liquid of lake up to 100 K than to cool it down to 80 K, temperature at which the fluid might solidify.

3 ESTIMATION OF THE SPEED OF SOUND IN A TERNARY MIXTURE OF NITROGEN, METHANE AND ETHANE

The speed of sound in a given substance is computed from its thermodynamic properties using the general equation (see for instance Diamantonis & Economou 2011)

$$u = \sqrt{\frac{C_P}{C_V} \left(\frac{\partial P}{\partial \rho} \right)_T} \quad (3)$$

with C_P and C_V respectively the isobaric and isochoric specific heat, P is the pressure, ρ the density and T the temperature. This equation can be easily derived from first principles and classical thermodynamics equations (*e.g.* Lobo & Ferreira 2006). The Helmholtz free energy is generally used in statistical thermodynamics to express EoS, since most properties of interest can be obtained by proper differentiation of it. With PC-SAFT, the total Helmholtz energy – denoted A – can be written as a sum

$$A = A^{(\text{id})} + A^{(\text{res})} \quad (4)$$

in which $A^{(\text{id})}$ represents the Helmholtz energy of the cor-

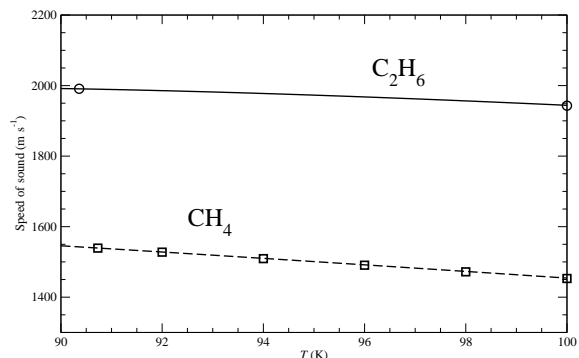


Figure 2. Comparison between experimental determination of the speed of sound (symbols) for CH₄ and C₂H₆ and the outputs of our model. Squares: experimental data from Setzmann & Wagner (1991), circles: data taken in Younglove & Ely (1987).

responding ideal-gas, $A^{(\text{res})}$ being the residual contribution that accounts for intermolecular interactions. In the PC-SAFT framework molecules are conceived to be chains comprised of freely jointed spherical segments. Inter-segment and intermolecular potentials are introduced and the theory provides the resulting Helmholtz energy of the macroscopic system. The reader who is interested in the nature of the terms included in $A^{(\text{res})}$ is invited to consult the vast literature devoted to the basis of PC-SAFT. The foundations of this theory have been originally published by Gross & Sadowski (2001). A good introduction to PC-SAFT can be found, in Soo (2011). This theory has been extensively tested in the context of cryogenic liquids by Tan et al. (2013) and it has also been proved that it reproduced laboratory data, particularly isotherms and binary diagrams, with a very satisfying degree of accuracy. However, despite its great performances, PC-SAFT, similarly to the theories applied by Cordier et al. (2009) or Glein & Shock (2013), include free parameters that have to be adjusted; but theories belonging to the “SAFT family” have the advantage to rely on a strong statistical physics basis. Moreover, I have checked that PC-SAFT reproduces the reference ternary mixture (N₂, CH₄, C₂H₆) of Gabis (1991) provided as supplementary data by Glein & Shock (2013).

In the frame of this EoS, each considered species is characterized by the PC-SAFT three parameters, namely the segment diameter σ , the depth of the potential ϵ/k_B , and the number of segments per chain m ; the values of those parameters have all been given in Tan et al. (2013); Cordier et al. (2016). Beside this, the binary interaction parameters k_{ij} ’s are also taken in this article. Eq. (5) can be rewritten as

$$u = \sqrt{\frac{C_P}{C_V} \frac{1}{k_T \rho}} \quad (5)$$

where $k_T^{-1} = \rho(\partial P/\partial \rho)_T$. The density ρ and the derivative $(\partial P/\partial \rho)_T$ are provided by PC-SAFT, whereas the non-ideal isobaric heat capacity of the mixture C_P is derived using the relation

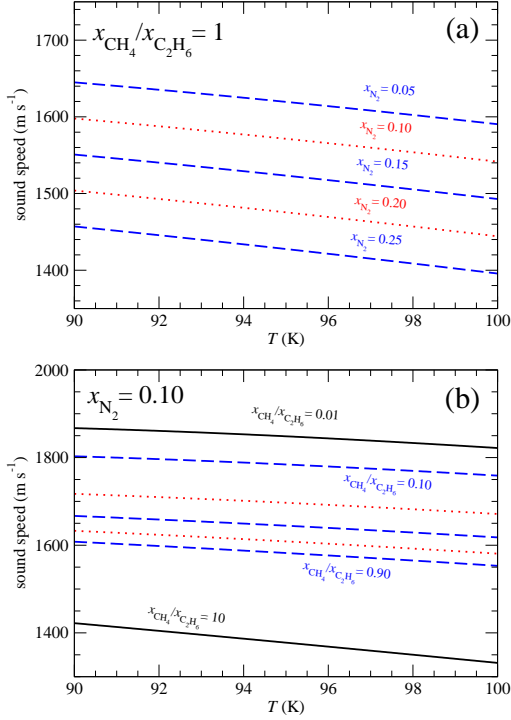


Figure 3. The calculated speed of sound as a function of temperature in the considered ternary mixture: (a) the ratio $r_{46} = x_{\text{CH}_4}/x_{\text{C}_2\text{H}_6}$ is fixed at 1 while the nitrogen mole fraction x_{N_2} takes the value 0.05, 0.10, 0.15, 0.20 and 0.25. (b) The mole fraction of nitrogen is set to 0.10 while the ratio $r_{46} = x_{\text{CH}_4}/x_{\text{C}_2\text{H}_6}$ takes the values 0.01, 0.10, 0.30, 0.50, 0.70, 0.90 and 10. For all calculations, the pressure has been fixed at 1.5 bar, the observed value on Titan’s surface (Neumann 2005).

$$C_P - C_V = \frac{T \alpha^2}{k_T \rho} \bar{M} \quad (6)$$

where $\alpha = k_T(\partial P/\partial T)_V$ is also computed with PC-SAFT. \bar{M} represents the average molar mass of the mixture. The isochoric heat capacity C_V is given by (see Diamantonis & Economou 2011, Eq. 19)

$$C_V = \underbrace{-T \left(\frac{\partial^2 A^{(\text{id})}}{\partial T^2} \right)_V}_{=C_V^{(\text{id})}} - T \left(\frac{\partial^2 A^{(\text{res})}}{\partial T^2} \right)_V \quad (7)$$

The term denoted $C_V^{(\text{id})}$ is the isochoric heat capacity of the corresponding ideal gas. I made a series of tests to evaluate this term. Among them, I used the group-contribution method developed by Joback and Reid (Joback 1984; Joback & Reid 1987) and summarized in Poling et al. (2007). This approach consists of an approximate estimation of the requested thermodynamical quantities. Unfortunately, doing so did not yield to speeds of sound in very good agreement with the tabulated experimental data. As a consequence, I found that adjusting the individual heat capacities $C_{P,i}^{(\text{id})}$ by fitting the individual velocity of sound data, gives

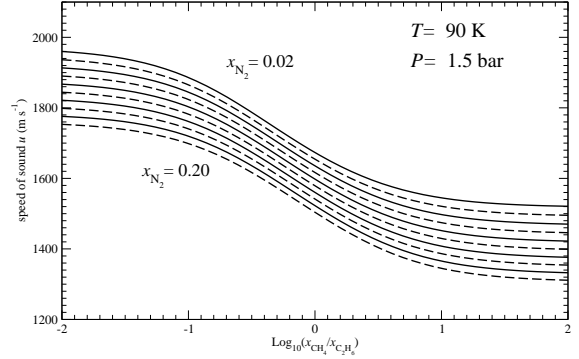


Figure 4. Tabulated speed of sound versus ratio $x_{\text{CH}_4}/x_{\text{C}_2\text{H}_6}$ at $T = 90$ K and $P = 1.5$ bar, for mole fraction of nitrogen $x_{\text{N}_2} = 2 \times 10^{-2}, 4 \times 10^{-2}, 6 \times 10^{-2}, 8 \times 10^{-2}, 1 \times 10^{-1}, 1.2 \times 10^{-1}, 1.4 \times 10^{-1}, 1.6 \times 10^{-1}, 1.8 \times 10^{-1}$ and 2×10^{-1} .

much better results. This isochoric $C_V^{(\text{id})}$ is derived from $C_{P,i}^{(\text{id})}$ thanks to Mayer’s law; while C_V comes from Eq. (7), Eq. (6) provides C_P . A comparison between the model outputs and experimental data is presented in Fig. 2. The agreement for methane speed of sound (Setzmann & Wagner 1991); and also for ethane data (Younglove & Ely 1987) appears very good. I have also been able to reproduce the velocity of sound in liquid nitrogen at 77 K (859 m s^{-1} , Zuckerwar & Mazel 1985). In Fig. 3 the calculated speed of sound for temperatures between 90 K and 100 K, is plotted for ranges of mole fractions that cover the plausible abundances of Titan’s seas. As it can be seen in panel (a) and (b) of this figure, u always decreases when T increases. It is noticeable that the amplitude of these variations, between the boundaries of the considered interval, is roughly $\sim 50 - 60 \text{ m s}^{-1}$. This implies an accuracy of measurements of a few percents, if we want to capture the variations of u with temperature. The curves in Fig. (3) do not exhibit local extrema, this is a useful (and expected) feature in the perspective of data inversion. Finally, we can remark that, not surprisingly, the computed speed of sound gets higher when the average molar mass increases.

4 DATA INVERSION

One major goal of this discussion is to assess the possibility of deriving chemical composition information from acoustic velocity. Thus, I have simulated a lake chemical composition extraction from artificial acoustic measurements. The samples were constructed using a finite number of sound velocities, computed with the model, for a list of N temperatures in the range of interest, *i.e.* 90–100 K. In general, an inverse problem, like the one we are facing here, can be treated in different ways (Aster et al. 2012). In a first attempt, I have tried to minimize a likelihood function based on a χ^2 . However, a very bad convergence was observed. Instead, I built pre-computed tables of speeds of sound depending on three parameters: the temperature T , the nitrogen mole fraction

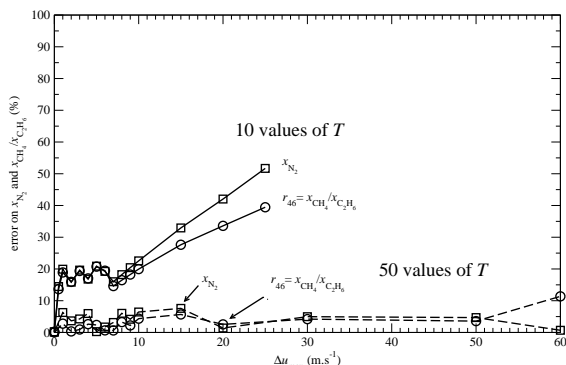


Figure 5. Results of simulations of “experimental” data inversion. Two sets of “experimental” data have been analyzed: one containing speeds of sound for 10 values of temperature uniformly distributed between 90 and 100 K, a second consisting in 50 values. In order to mimic instrumental errors on speed of sound measurements, a random signal has been added to the data, originally computed using my PC-SAFT-based thermodynamic model. This signal consists of randomly chosen speed of sound contributions uniformly distributed between $-\Delta u_{\max}$ and $+\Delta u_{\max}$, where Δu_{\max} is the maximum measurement error allowed in the simulation. This plot represents the error on chemical composition determination as a function of Δu_{\max} .

x_{N_2} and the ratio $r_{46} = x_{CH_4}/x_{C_2H_6}$. The data inversion is performed using the following algorithm:

(i) for each “experimental” temperature T_i , the value of the ratio r_{46} is searched by solving the equation $u_{\exp} = u_{\text{Table}}$, for each value of x_{N_2} implemented in the multidimensional table. In Fig. 4, I have displayed examples of curves giving u versus $\log_{10}(r_{46})$ for a temperature fixed at 90 K and a nitrogen mole fraction ranging from 0.02 to 0.20. This way, one builds a set of curves providing $x_{N_2}^{(T)}$ as a function of the ratio r_{46} .

(ii) in a second step, for each couple of temperature values $(T_i, T_j)_{i \neq j}$, the coordinates of the intersection point of the curves $x_{N_2}^{(T_i)}(r_{46})$ and $x_{N_2}^{(T_j)}(r_{46})$ are determined using a root finding method. It should be noted that, in an ideal situation, all the curves have to intersect strictly at the same point $(r_{46}|_0, x_{N_2}|_0)$ characterizing the chemical composition of the studied liquid. In practice this is not the case, due to numerical errors, and above all because of experimental uncertainties, the derived points are scattered around an average position. The barycenter of these points represents the observed chemical composition, while the scattering offers information on uncertainties associated with this derivation.

Here, the uncertainties were estimated by computing the difference between the “true” composition used at the time of the look-up-table construction, and the abundances inferred with our inversion algorithm. It is valuable to estimate the sensitivity of the results (*i.e.* derived values of the composition of the liquid) to the uncertainties on the measured speed of sound. In order to mimic the instrumental errors, I added a random signal to velocity values, originally computed with the thermodynamical model. Subsequently,

the analysis of these surrogate samples is done by the use of the algorithm described above. Thus, I applied artificial speed errors Δu_i uniformly distributed between $-\Delta u_{\max}$ and $+\Delta u_{\max}$, where Δu_{\max} is an arbitrary chosen value, representing the maximum error allowed in the numerical test. As already noticed in Sect. 3, the accuracy of the speed of sound in the considered liquid must be much lower than $\sim 50 - 60 \text{ m s}^{-1}$ for a typical value of speed around 1500 m s^{-1} . Then the required instrumental precision should be a priori better than $\sim 50 \text{ m s}^{-1}$. In Fig. 5 we have reported the results of two simulations: one based on a set of $N = 10$ values of temperature uniformly distributed between 90 and 100 K, the second consisting in $N = 50$ values. In this figure, the errors on nitrogen mole fraction and errors on the ratio $r_{46} = x_{CH_4}/x_{C_2H_6}$ were plotted as functions of the value of the maximum error Δu_{\max} . Both simulations were done using an initial “true” chemical composition corresponding to $x_{N_2} = 0.10$ and $x_{CH_4}/x_{C_2H_6} = 0.50$. As one can see, for $N = 10$ the errors are relatively stable around $\sim 20\%$ for Δu_{\max} up to $\sim 10 \text{ m s}^{-1}$; beyond this value the errors increase regularly and finally the algorithm no longer converges properly for $\Delta u_{\max} \sim 25 \text{ m s}^{-1}$. This numerical experiment corresponds to a temperature control at a 1-K level, which appears well feasible. As a corollary, in that case, the speed of sound measurements have to be done with an absolute accuracy better than 10 m s^{-1} , which corresponds to a relative precision better than 10^{-2} . In Fig. 5, the $N = 50$ numerical experiment shows clearly that, for a given Δu_{\max} , the increase of the number of measurements yields to an appreciable improvement in the chemical composition determination. In addition, the results of the inversion remains acceptable (errors remain below $\sim 10\%$) up to $\Delta u_{\max} \sim 60 \text{ m s}^{-1}$. Nonetheless, a number of measurements of $N = 50$ requires a temperature control of the liquid at the level of $\sim 0.2 \text{ K}$ that is more difficult to achieve than a $\sim 1 \text{ K}$ level. This gives an idea of the precision needed on the temperature control. The dependency on N is anticipated since the final step of the inversion algorithm consists in an average. During the design stage of the instrument, a compromise will have to be found between temperature control and the accuracy of speed of sound determinations.

I have also checked that the errors on temperature have a negligible influence. For instance, in the case of a sample of 10 measurements between 90 and 100 K, randomly distributed errors on T with a maximum of $\pm 0.2 \text{ K}$ lead to errors on speed of sound below 0.8 m.s^{-1} while a maximum error of $\pm 0.5 \text{ K}$ produces speed uncertainties not larger than 2 m.s^{-1} , value well below the Δu_{\max} values considered above. This demonstrates that, if the temperature accuracy required by a number of independent measurements can be guaranteed (*e.g.* better than 1 K for 10 measurements between 90 and 100 K), then the effects of the inaccuracies in the temperature measurements on the results are negligible compared to the uncertainties in the speed of sound measurements.

Finally, photochemical models of Titan’s atmosphere show that propane could be produced (Lavvas et al. 2008a,b). Thus, I introduced some amount of propane in the case where Δu_{\max} is fixed at 30 m s^{-1} . The chemical composition of the “experimental sample”, *e.g.* $x_{CH_4} = 0.30$,

$x_{\text{N}_2} = 0.10$ and $x_{\text{C}_2\text{H}_6} = 0.60$, is replaced by $x_{\text{CH}_4} = 0.30$, $x_{\text{N}_2} = 0.10$, $x_{\text{C}_2\text{H}_6} = 0.55$ and $x_{\text{C}_3\text{H}_8} = 0.05$. These data are then used in the algorithm which assumes a sample composed only by the ternary mixture (CH_4 , N_2 , C_2H_6). I found that, even with such a small amount of propane, the derived abundances of N_2 and the ratio r_{46} are substantially affected: the error on x_{N_2} is around 20% (instead of $\sim 5\%$ without C_3H_8) while r_{46} presents an error of about 100%. This numerical test emphasizes the sensibility of the velocity of sound to composition. Obviously, to overcome this issue, the best solution is to introduce the measurement of another independent physical quantity.

5 CONCLUSION

In this work, I shown that the use of a realistic model, based on PC-SAFT, prevents the appearance of unphysical situations, like a speed of sound discontinuity, already noticed in previous published papers (Hagermann et al. 2005; Arvelo & Lorenz 2013). However, even such sophisticated models need to be constrained by empirical data. Consequently, new laboratory measurements of sound speeds would be greatly useful, particularly in the case of mixtures. The simultaneous measurements of refractive index, density, thermal conductivity, electromagnetic permittivity and speed of sound required by Hagermann *et al.*'s method is not easy to achieve and requires a complex set of sensors. Alternatively, the dynamic method proposed here, whereby the speed of sound is measured at different temperatures; provides a better composition estimation with accuracy comparable to that of the measured physical quantities (*i.e.* a few percents), here the speed of sound. Indeed, with only 50 temperature measurements, I have shown that the errors in derived composition remain below 10%. I also emphasized that the method employed by Hagermann et al. (2005) implicitly assumes the existence of a device implementing a temperature control by heating since they need thermal conductivity determinations. This fact suggest that, the two approaches could be combined in a future mission concept.

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